

ATTACHMENT A

## Patent claims:

- 1.(original) Organic solutions containing metal(IV) salts and oxoacids of phosphorus from which, after evaporation of the solvent, insoluble compounds of general composition  $M(IV)(O_3P-G)_{2-n}(O_3P-R^1-X)_n$  can be obtained, where M(IV) is a tetravalent metal, -G is a generic inorganic or organic group, -R<sup>1</sup>- is an organic group, -X is an acid group and n is a coefficient ranging from 0 to 1.5.
2. (original) Organic solutions of claim 1 wherein the anion of the tetravalent metal salt is preferably chosen among carboxylates, chlorides and alkoxides.
3. (currently amended) Organic solutions of claim 1 or 2 wherein the tetravalent metal salt is preferably chosen between Zr, Ti, Sn and Ce or their mixture.
4. (currently amended) Organic solutions of ~~any of claims 1-3~~ claim 1 wherein the tetravalent salt is preferably the zirconyl propionate or chloride.
5. (currently amended) Organic solutions of ~~any of the preceding claims~~ claim 1 wherein the group -G is preferably chosen among the acid groups -OH ; -R<sup>2</sup>-SO<sub>3</sub>H and -R<sup>2</sup>-PO<sub>3</sub>H<sub>2</sub>, where -R<sup>2</sup>- is an organic group with preferably linear chain such as -(CH<sub>2</sub>)<sub>m</sub>- and -(CF<sub>2</sub>)<sub>m</sub>-.
6. (currently amended) Organic solutions of ~~any of the preceding claims~~ claim 1 wherein the group -R<sup>1</sup>- is an arylene group chosen preferably among -C<sub>6</sub>H<sub>4</sub>-; -C<sub>6</sub>H<sub>4</sub>-CH<sub>2</sub>- and -C<sub>6</sub>H<sub>4</sub>-CF<sub>2</sub>-.
7. (currently amended) Organic solutions of ~~any of the preceding claims~~ claim 1 wherein the acid group -X is chosen between -SO<sub>3</sub>H, -PO<sub>3</sub>H<sub>2</sub> and -COOH.

8. (currently amended) Organic solutions of ~~any of the preceding claims~~ claim 1 wherein the organic solvent is chosen among the protonable solvents, especially N,N-dimethylformamide, N-methyl-2-pyrrolidone, dioxane, dimethylsulfoxide, acetamide, acetonitrile, various alkanols and/or their mixtures, commonly used for dissolving the proton conducting ionomers of the state of art.
9. (currently amended) Use of the organic solutions of ~~any of the preceding claims~~ claim 1 for the insertion of nano-particles of tetravalent metal salts, preferably phosphate-phosphonates, within the pores of polymeric or inorganic porous membranes.
10. (currently amended) A method for the filling of porous membranes of claim 9 with tetravalent metal salts, especially phosphate-phosphonates, based on the following steps:
  - a) preparation of the organic solution ~~of claims 1-8~~ which, at the same time, may also contain a polymer and/or an ionomer of the state of the art;
  - b) impregnation of the porous membrane with such a solution;
  - c) elimination of the solvent;
  - d) repetition of the steps b and c until the wished percentage of pore filling is obtained.
11. (currently amended) Proton conducting composite membranes made of polymeric or inorganic porous membranes with pores filled with tetravalent metal salts, especially phosphate-phosphonates, or mixtures of said compounds with a proton conducting ionomer and especially prepared by using the solutions of ~~claims 1-8~~ claim 1.
12. (original) Proton conducting composite membranes of claim 11 wherein the polymeric porous membrane is preferably chosen between those made of chemically and/or thermally stable polymers, especially

polytetrafluoroethylene (PTFE), polyvinylidene fluoride (PVDF),  
polyesters, polyethersulfones and fluoroelastomers.

13. (currently amended) Proton conducting composite membranes ~~of any of claims 11 or 12~~ claim 11 wherein the pore dimensions of the porous membranes are preferably in the range 0.02-20  $\mu\text{m}$ , especially 0,1-10  $\mu\text{m}$ , preferably 0,4-2  $\mu\text{m}$  and the porosity >10 %, especially >50 %, preferably 65-90%.
14. (currently amended) Proton conducting composite membranes of any ~~of claims 11-13~~ claim 11 wherein the tetravalent metal salts, preferably phosphatephosphonates, for the filling of pores are chosen between  $\text{Zr}(\text{O}_3\text{P-CH}_2\text{-PO}_3\text{H}_2)_2$  and compounds of the series  $\text{Zr}(\text{O}_3\text{P-OH})_{2-n}(\text{O}_3\text{P-C}_6\text{H}_4\text{-SO}_3\text{H})_n$ , and  $\text{Zr}(\text{O}_3\text{P-C}_6\text{H}_4\text{-SO}_3\text{H})_{2-n}(\text{O}_3\text{P-CH}_2\text{-PO}_3\text{H}_2)_n$ , with n in the range 0.1-1.5.
15. (currently amended) Composite membranes made up of a porous ceramic membrane partially filled with a tetravalent metal salt, preferably phosphatephosphonate, according to ~~one of claims 1-7~~ claim 1 exhibiting catalytic activity.
16. (currently amended) Composite membranes ~~of claim 15~~ made up of a porous ceramic membrane partially filled with a tetravalent metal salt, preferably phosphatephosphonate, wherein the tetravalent metal salts, preferably phosphate-phosphonate, with catalytic activity is chosen among those reported in claim 14.
17. (currently amended) Use of the organic solutions of ~~any of claims 1-8~~ claim 1 for the preparation of nano-polymers constituted by nano-particles of tetravalent metal salts, preferably phosphate-phosphonates, dispersed in the matrix of organic or inorganic polymers soluble in the same solvents.

18. (currently amended) Use of the organic solutions of ~~any of claims 4-8~~ claim 1 for the preparation of nano-polymers of ~~claim 17~~ constituted by nano-particles of tetravalent metal salts, preferably phosphate-phosphonates, dispersed in the matrix of organic or inorganic polymers soluble in the same solvents, wherein the organic polymeric matrix is that of a proton conducting ionomer.
- 19.(currently amended) A method for the preparation of nano-polymers and nano-ionomers ~~of any of claims 17-18~~ constituted by nano-particles of tetravalent metal salts, preferably phosphate-phosphonates, dispersed in the matrix of organic or inorganic polymers soluble in the same solvents based on:
- a) preparation of an organic solution having one of the compositions reported in the any of claims 1-8 and at the same time containing a polymer and/or an ionomer of the state of the art;
  - b) elimination of the solvent.
20. (original) The method for the preparation of nano-polymers and nanoionomers of claim 19 wherein the elimination of the solvent is preferably performed by evaporation or with a non- solvent of the polymer or ionomer.
21. (original) Nano-polymers constituted by nano-particles of tetravalent metal salts, preferably phosphate-phosphonates, dispersed in the matrix of organic or inorganic polymers.

22. (original) Nano-polymers of claim 21 wherein the polymeric matrix is that of a synthetic ionomer of the state of the art preferably chosen among perfluorosulphonic polymers, sulfonated polyetherketones (sPEK), sulfonated polyethersulfones and sulfonated polyvinylidenefluoride (sPVDF).

23. (currently amended) Nano-polymers of claims 21 ~~or 22~~ wherein the nano-particles of tetravalent metal salts, preferably phosphate-phosphonates, dispersed in the polymeric matrix are chosen among those exhibiting proton conductivity  $> 10^{-2} \text{ S cm}^{-1}$  at 70 °C and 95% relative humidity.

24. (currently amended) Nano-polymers of ~~any of claims 21-23~~ claim 21 wherein the nano-particles of tetravalent metal salts, preferably phosphate-phosphonates, are those constituted by the same compounds of claim 14.

25. (currently amended) Use of the organic solutions of ~~any of claims 1-8~~ claim 1 for the preparation of membranes constituted by nano-polymers of claims 21-24.

26. (currently amended) Nano-ionomeric proton conducting membranes constituted by nanopolymers of ~~any of claims 21-25~~ claim 21.

27. (currently amended) Use of the organic solutions of ~~any of claims 1-8~~ claim 1 for an easy insertion of a large variety of lamellar nano-particles of tetravalent metal salts, preferably phosphate-phosphonates, in the membrane/electrode interfaces of PEM FCs.

28. (currently amended) Use of the organic solutions of ~~any of claims 4-8~~ claim 1, with the addition of ionomers and/or other proton conducting compounds soluble in the same solvents, for an easy insertion of a large variety of lamellar nano-particles of tetravalent metal salts, preferably phosphate-phosphonates, in mixture with other proton conducting compounds in the membrane/electrode interfaces of PEM FCs.
29. (currently amended) Use of proton conducting membranes constituted by inorganic or polymeric porous membranes with pores filled with tetravalent metal salts, preferably phosphate-phosphonates, ~~of any of claims 11-16~~ claim 11 and of membranes constituted by nano-polymers ~~of claim 26~~ nano-particles of tetravalent metal salts, preferably phosphate-phosphonates, dispersed in the matrix of organic or inorganic polymers, in electrochemical devices.
30. (original) Use of proton conducting membranes of claim 29 in electrochemical devices specifically planned for generating electrical energy from the oxidation of a fuel.
31. (original) Use of proton conducting membranes of claim 29 in fuel cells specifically planned for electrical vehicles and/or for portable electrical devices.
32. (original) Use of composite membranes of claim 29 for improving the global performance of ionomeric membranes of the state of the art in hydrogen, indirect methanol and direct methanol fuel cells.
33. (currently amended) Use of membranes of claim 11, ~~15 and 16~~ in catalytic membrane reactors.